

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

IN RE APPLI-
CATION OF : K. Lassila, et al.

SERIAL NO. : 09/427,317

FILED : 26 October 1999

FOR : Malic Acid Diester Surfactants



GRP. ART UNIT: 1714

EXAMINER: P. Michl

Assistant Commissioner for Patents
Washington, D.C. 20231

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Sir:

RULE 132 DECLARATION

I, Caroline S. Slone, residing at 1605 Sleepy Hollow Road, Quakertown, PA 18951 hereby declare as follows:

1. I am employed by Air Products and Chemicals, Inc., the assignee of the invention described in the above-identified Application. I have been employed by Air Products and Chemicals, Inc. for 3 years working the past 2 years in the Specialty Additives Group of the Performance Chemicals Division.

2. I am a co-inventor of the subject matter claimed in the above-identified application.

3. I have read and understand the Mattai patent (US 5,904,917), which was relied upon by the Examiner in rejecting the claims pending in the above-identified Application.

4. Under my supervision and direction, dynamic surface tension data and foaming data were collected for oil components suggested for use in the sun protection compositions of Mattai. Table 1 presents comparative dynamic surface tension data for dioctylmalate and diethylmalate. The data were acquired for aqueous compositions obtained by the addition of the oil component to water. The experiments were performed to assess the inherent dynamic surface tension reducing ability of the additives. Examination of these materials in water was necessary as Mattai's sun protection compositions contain surfactants, such as AEOS 2EO, which are also capable of reducing surface tension. Moreover, it is impractical to measure the dynamic surface tension of viscous lotions such as Mattai's compositions.

5. Diethylmalate was purchased from TCI America and used as received. Dioctylmalate was prepared by esterification of malic acid with 1-octanol as shown in Paragraph 6. All materials studied ranged from >94% to >99% pure.

6. Di-octyl-DL-malate Synthesis. Di-octyl-DL-malate was prepared using p-toluenesulfonic acid as the catalyst. To the reaction flask, DL-malic acid (80.14 g), 1-octanol (311.33 g) and p-toluenesulfonic acid (11.37 g) were added. The mixture was placed under nitrogen and heated to reflux. At 109°C, two phases started to collect in the Dean-Stark trap. The reaction was maintained at 110-120°C for 1 hr and 20 min and then heated to 135°C over 2 hr. During this time, the water from the water/alcohol azeotrope was removed. The product was dissolved in diethyl ether, washed with saturated sodium bicarbonate (200 mL) 2 times, washed once with water, and dried over magnesium sulfate. Diethyl ether was removed via rotary evaporation. Di-octyl-DL-malate was obtained as a low viscosity, slightly hazy colorless liquid with no detectable odor (185.42 g, 87% yield) after removing the residual 1-octanol via vacuum distillation.

7. Dynamic Surface Tension Measurements. Dynamic surface tension data were obtained for aqueous solutions of dioctylmalate and diethylmalate using the maximum bubble pressure method at bubble rates from 0.1 bubbles/second (b/s) to 20 b/s. The maximum bubble pressure method of measuring surface tension is described in Langmuir 1986, 2, 428-432. These data provide information about the performance of a surfactant at

conditions from near-equilibrium (0.1 b/s) through extremely high surface creation rates (20 b/s).

8. Dioctylmalate was insoluble at both 0.001 wt% and 0.1 wt% and therefore the dynamic surface tension measurements were made in Examples A and B on the saturated aqueous solutions after removal of the insolubles. Diethylmalate was completely soluble at 5 wt% for Example C.

Table 1

| Malate | Ex | Wt% | γ (0.1 b/s) | γ (1 b/s) | γ (6 b/s) | γ (20 b/s) |
|---------|----|-------|--------------------|------------------|------------------|-------------------|
| Dioctyl | A | 0.001 | 70.3 | 70.6 | 71.0 | 72.4 |
| Dioctyl | B | 0.1 | 51.8 | 68.6 | 70.5 | 72.6 |
| Diethyl | C | 5 | 46.1 | 46.5 | 47.0 | 48.6 |

γ = dynes/cm

9. Foaming Measurements. The foaming properties of 0.1 wt% solutions of the dialkylmalates were tested using a procedure based upon ASTM D 1173 - 53. In this procedure, a 0.1 wt% solution of the malate is added from an elevated foam pipette to a foam receiver containing the same solution. The foam height is measured at the completion of the addition ("Initial Foam Height") and the time required for the foam to dissipate at the air-liquid interface ("Time to 0 Foam") is recorded. The foaming data are presented in Table 2.

Table 2

| Malate | Ex | Wt% | Initial foam (cm) | Time to zero foam (sec) |
|---------|----|-----|-------------------|-------------------------|
| Dioctyl | B | 0.1 | 0 | 0 |
| Diethyl | C | 0.1 | 2.4 | 7 |

10. Since dioctylmalate was insoluble at 0.1 wt%, the foam test was performed on the soluble fraction of a formal 0.1 wt% solution. Dioctylmalate did not generate any foam. However, diethylmalate generated 2.4 cm of foam compared to 1.3 cm of foam generated by

diisopropylmalate which is shown as Example 21 in Table 2 of the above-identified application. The difference between the diisopropyl and diethyl malates in terms of the time to zero foam is not significant in practice. It is more important to avoid foam formation initially.

I hereby declare further that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that the statements are made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Carole S. Slone

Caroline S. Slone

February 26, 2001

Date